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Tammeorg, Olga

2018-05

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Tammeorg , O , Haldna , M , Noges , P , Appleby , P , Mols , T , Niemisto , J , Tammeorg , P & Horppila , J 2018 , ' Factors behind the variability of phosphorus accumulation in Finnish lakes ' , Journal of Soils and Sediments , vol. 18 , no. 5 , pp. 2117-2129 . <https://doi.org/10.1007/s11368-018-1973-8>

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<http://hdl.handle.net/10138/307852>

<https://doi.org/10.1007/s11368-018-1973-8>

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**Factors behind the variability of phosphorus accumulation in Finnish lakes**

**Olga Tammeorg<sup>1,2</sup> • Marina Haldna<sup>2</sup> • Peeter Nõges<sup>2</sup> • Peter Appleby<sup>3</sup> • Tõnu Möls<sup>2</sup> • Juha Niemistö<sup>1</sup> •  
Priit Tammeorg<sup>4</sup> • Jukka Horppila<sup>1</sup>**

<sup>1</sup>Department of Environmental Sciences, University of Helsinki, P.O. Box 65, 00014, Helsinki, Finland

<sup>2</sup>Centre for Limnology, Estonian University of Life Sciences, 61117 Rannu, Tartumaa, Estonia

<sup>3</sup>Department of Mathematical Sciences, University of Liverpool, L69 3BX, Liverpool, UK

<sup>4</sup>Department of Agricultural Sciences, University of Helsinki, P.O. Box 27, FI-00014, Helsinki, Finland

✉ Olga Tammeorg

[olga.tammeorg@helsinki.fi](mailto:olga.tammeorg@helsinki.fi)

## Abstract

*Purpose.* Phosphorus retention ( $TP_{acc}$ ) is one of the major water quality regulators in lakes. The current study aimed at ascertaining the specific lake characteristics regulating  $TP_{acc}$ . Moreover, we were interested whether NAO (North Atlantic Oscillation), a proxy of climatic forcing, can explain variability in  $TP_{acc}$ , additionally to that ascribed to lake characteristics.

*Materials and methods.* Sediment cores were obtained from 21 Finnish lakes, subject to radiometric dating and measurements of TP concentrations. Principal components (PCs) were generated using lake characteristics that are usually included into the modelling of  $TP_{acc}$  (e.g., lake area, lake depth, catchment area, P inflow), but also the parameters that the classical models usually missed (e.g., anoxic factor). We used significant principal components (PCs), specific combinations of lake characteristics and monthly NAO values as predictors of  $TP_{acc}$ .

*Results and discussion.* Lake characteristics explained the bulk of  $TP_{acc}$  variability. The most influential factors (positive drivers) behind  $TP_{acc}$  included PC1 (representing mainly deep lakes), PC2 (small lakes with high levels of anoxia and water column stability), PC3 (productive lakes, with large catchment area and short water residence time), PC4 (lakes with high water column stability, low anoxic factor, and relatively high sediment focusing), and PC5 (lakes with high levels of P inflow, anoxia and long water residence time). Additionally, we found a potential negative effect of NAO in October on the annual  $TP_{acc}$ . This NAO was significantly positively related to temperatures in surface and near-bottom water layer (also their difference) in autumn, suggesting the possible implications for the internal P dynamics. Increased mineralization of organic matter is the most likely explanation for the reduced  $TP_{acc}$  associated with NAO driven water temperature increase.

*Conclusions.* The analysis presented here contributes to the knowledge of the factors controlling P retention. Moreover, this spatially and temporally comprehensive sediment data can potentially be a valuable source for modelling climate change implications.

**Keywords** Lake characteristics • Lakes • NAO • Phosphorus accumulation rate • Phosphorus retention

## 1 Introduction

Being one of the major regulators of the productivity in waterbodies, the phosphorus (P) retention has been in the focus of aquatic ecosystem modelling for about half of a century. In the mass balance models, the P retention is often determined as the difference between the inflowing and outflowing P. As an alternative that enables to eliminate the need for extensive monitoring programmes, the net P retention can be estimated by multiplying the net sediment accumulation rate with its P content ( $TP_{acc}$ ; Dillon and Evans 1993; Boers et al. 1998). There have been many attempts to predict P retention from a number of characteristics (e.g., Dillon and Kirchner 1975; Larsen and Mercier 1976; Vollenweider 1975), whereby the most common predictors of the P retention include phosphorus and hydraulic loading rate, TP particle settling velocity and mean depth. Nevertheless, the large prediction errors were found to be associated with those models, as these do not account for P release (Nürnberg, 1984). Sediments can serve as an important source of P in the years following reduction of external loading until the legacy P pool is reduced or buried in the deeper sediments (Sas 1990; Jeppesen et al. 2005; Søndergaard et al. 2013). Moreover, Benjamin and Brett (2008) demonstrated that the prevailing approach of conceptualization of the P retention overestimates the impact of the parameters usually used. The authors found that the best mass balance model tested could explain 84% of the variability in log-transformed lake TP concentrations, while it explained only 35% of the variability in TP retention and resulted in a large prediction error for individual lakes. The complex coupling of sediment composition, external load, catchment hydrology, lake morphometry, and biogeochemical reactions has been recognized to control P retention (Hupfer and Lewandowski 2008; Søndergaard et al. 2013; Huser et al. 2016). Hence, there is still a need for a P retention model that could better account for the lake specifics.

Climate change can affect P retention via variations in air temperature and precipitation that both influence P transport to the lakes (Jeppesen et al. 2009; Pettersson et al. 2010). Additionally, changes in temperature and wind have considerable implications for the vertical transport of P in lakes (Spears and Jones 2010; Tammeorg et al. 2013; Tammeorg et al. 2016; Woolway et al. 2017). Generally, climate change is associated with increased net P accumulation in lakes due to enhanced external nutrient loading leading also to increased internal P loading (Jeppesen et al. 2011). The North Atlantic Oscillation (NAO) index has performed as a good indicator of climatic forcing in European lakes. NAO index has shown to have a positive correlation with e.g. water temperatures, some lake water chemistry variables (Blenckner et al. 2007), wind speed (Vermaat et al. 2008), particularly in winter and spring, and wave-mixed depths (Spears and Jones 2010). Nevertheless, there is still a lack of knowledge on the relationship of NAO with P retention. As sediment records can reflect climatic variability (Bennion et al. 2006;

Rose et al. 2010; Sánchez-López et al. 2016) connecting  $TP_{acc}$  to climatic variation via NAO could be a useful tool to target that knowledge gap.

In the current study, we aimed at ascertaining the specific combinations of lake characteristics, principal components (PCs) that determine  $TP_{acc}$ , using data obtained from dated sediment cores collected from 21 Finnish lakes. Principal components (PCs) were generated using lake characteristics that are usually included into the modelling of TP retention (e.g., area and depth of lakes, size of catchment area and P inflow), but also the parameters missed by the classical models (e.g., anoxic factor, Osgood's index). Additionally, we coupled this information and the data on NAO for the time period covered by the sediment cores to elucidate the potential role of climatic variability in  $TP_{acc}$ . As climate change is primarily associated with changes in air temperature, which are closely coupled to water temperature, we were particularly interested whether potential NAO effects on  $TP_{acc}$  can be attributed to the temperature changes.

## 2 Methods

### 2.1 Study area

The 21 lakes of the study were all located in southern Finland, with their areas ranging from 0.25 to 155 km<sup>2</sup>. The mean depth of the lakes varied from 1.3 to 21.0 m (Table 1), and the maximum depth from 3 to 68 m. Most of the lakes had deep areas, which undergo periodic anoxia, generally in winter and during thermal stratification in summer. The values of the anoxic factor (i.e., the product of the duration of anoxia and the percentage of the anaerobic areas) varied from 0 for the nonstratifying lakes (Nürnberg 1984) to 50 d y<sup>-1</sup> (Tammeorg et al. 2017). The monitoring data (Finnish Environment Institute) indicated the trophic status ranging from mesotrophic to hypertrophic (Table 1). Mean phosphorus inflow,  $TP_{in}$  varied from 104 mg m<sup>-2</sup> y<sup>-1</sup> in mesotrophic lakes to 910 mg m<sup>-2</sup> y<sup>-1</sup> in (hyper)eutrophic lakes (Tammeorg et al. 2017). The catchments of the eutrophic and hypertrophic lakes have mainly been impacted by agricultural activities (Ekholm and Mitikka 2006). All studied lakes were subject to a variety of restoration methods (including wastewater diversion, biomanipulation, artificial aeration) during past 30 years (Table 1).

100 TP accumulation rate ( $TP_{acc}$ ,  $mg\ m^{-2}\ y^{-1}$ ) was calculated by multiplying the concentration of TP in the sediment  
101 layer by the sedimentation rate. For that, sediment cores were collected with HTH gravity corer from the deepest  
102 site of the lakes (the Kajaanselkä basin of Lake Vesijärvi was sampled at a site close to the maximally deep due to  
103 technical reasons) targeting the accumulation areas (Håkanson and Jansson 1983) in March 2013 and 2014, when  
104 the lakes were ice-covered. Sampling at locations that were predominantly stratified and anoxic during summer  
105 ensured also minimal core disturbances due to wind activity (sediment resuspension), and bioturbation. Low water  
106 temperatures during sampling lowered the risk of temperature-dependent transformations (e.g. P release) at the  
107 sediment-water interface. Moreover, as it was identified by visual inspection, sediment surface was oxidized  
108 inhibiting the release of P in the lakes studied. Dissolved oxygen concentration in the near-bottom water layer was  
109 mainly above  $7.0\ mg\ l^{-1}$ . Each of the cores was sectioned into 0.5 cm slices to a depth of 20 cm to cover the period  
110 for which also TP concentrations in the surface water layer and water temperature data were available, i.e. most  
111 recent three decades (1986-2014). All sediment samples (40 samples per lake) were freeze-dried and ground. The  
112 TP concentrations from the sediment subsamples were further determined using the methods by Koroleff (1979;  
113 Lachat autoanalyzer, QuickChem Series 8000; Lachat instruments, Loveland, USA) after wet digestion with  
114 sulphuric acid and hydrogen peroxide (Milestone Ethos 1600 microwave oven; Milestone, Sorisole, Italy).

115 Sedimentation rates were determined by dating cores (40 layers per core) by  $^{210}Pb$  and  $^{137}Cs$ . The analysis was  
116 performed at the Liverpool University Environmental Radioactivity Laboratory. Sub-samples from each core were  
117 analysed for  $^{210}Pb$ ,  $^{226}Ra$ , and  $^{137}Cs$  by direct gamma assay using Ortec HPGe GWL series well-type coaxial low  
118 background intrinsic germanium detectors (Appleby et al. 1986).  $^{210}Pb$  was determined via its gamma emissions  
119 at 46.5 keV, and  $^{226}Ra$  by the 295 keV and 352 keV  $\gamma$ -rays emitted by its daughter radionuclide  $^{214}Pb$  following 3  
120 weeks storage in sealed containers to allow radioactive equilibration.  $^{137}Cs$  was measured by its emissions at 662  
121 keV. The absolute efficiencies of the detectors were determined using calibrated sources and sediment samples of  
122 known activity. Corrections were made for the effect of self-absorption of low energy  $\gamma$ -rays within the sample  
123 (Appleby et al. 1992).  $^{210}Pb$  dates were calculated mainly using CRS model (Appleby and Oldfield 1978). Since  
124 in many cases the  $^{210}Pb$  record spanned no more than around three decades, the calculation of reliable dates  
125 demanded the use of well-defined  $^{137}Cs$  dates as reference points. The method is described in detail by Appleby  
126 (2001).

To quantify the possible impact of sediment focusing at the sampling area, a well-recognized issue (e.g., Eisenreich et al. 1989; Blais and Kalff 1993; Rowan et al. 1995; Lamborg et al. 2002; Heathcote and Downing 2014), focusing factors (FFs), calculated as the ratio of the measured mean  $^{210}\text{Pb}$  supply rate (flux) at the core site to the atmospheric flux, were determined for each site. The atmospheric flux (based on fallout data from a number of European sites and records in 36 Finnish cores held in the Liverpool University ERRC base) was estimated to be  $100 \pm 20 \text{ Bq m}^{-2} \text{ y}^{-1}$ . Although causes of high  $^{210}\text{Pb}$  supply rates at specific sites can also include allochthonous inputs of fallout  $^{210}\text{Pb}$  from the catchment (Appleby 2001), this was not thought to be a significant issue in our study, since our cores were collected from sites far from inlet streams at the deepest points of the lake.

Data on water temperature and surface water TP concentrations covering two-three most recent decades in the lakes studied were obtained from Hertta database (Finnish Env. Inst.). Besides air temperature, water temperatures can be affected by wind; thus, we analyzed also wind data. However, the more direct effects of the wind activity (through enhanced sediment resuspension at shallow areas and increased horizontal transport of these sediments to lake deeps, focusing) cannot be ignored. Data on wind speed for the Helsinki-Vantaa airport (8 measurements per day) were obtained from the Finnish Meteorological Institute. Daily NAO values were obtained from: <http://www.cpc.ncep.noaa.gov/products/precip/CWlink/pna/nao.shtml>. Studied (hydro)meteorological variables were averaged over the months from January to December for the years 1970–2014. Data on TP concentration in the lake water of the surface layer was used to reflect the trophic state history of the lakes.

### 2.3 Statistical methods

Raw NAO, water temperature, and wind speed data statistics are shown with boxplot diagrams. The trends in rates of sediment accumulation, TP concentrations in the sediments and  $\text{TP}_{\text{acc}}$ , and TP concentration in the surface water layer over the years 1986–2014 for the studied lakes were tested with linear regression analysis.

To ascertain lake characteristics responsible for the  $\text{TP}_{\text{acc}}$ , the Principal Component Analysis (PCA) was carried out. Principal components (PCs) were obtained as weighted linear combinations of the original variables. Original variables included those lake characteristics that were demonstrated to be of paramount importance for controlling lake phosphorus dynamics, i.e. maximum depth ( $D_{\text{max}}$ ), mean depth ( $D$ ), ratio of  $D_{\text{max}}$  to  $D$  (to represent the potential importance of lateral sediment flux, i.e. sediment focusing), lake area (LA), catchment area (CA), ratio of the CA to LA (used as a proxy of water residence time), inflow of P ( $\text{TP}_{\text{in}}$ ), anoxic factor (AF, to represent sediment P release due to anoxia), Osgood's index, or  $D \times \text{LA}^{-0.5}$  (OI, to represent water column stability).  $D_{\text{max}}/D$

correlated well with the focusing factor ( $r = 0.600$ ,  $p = 0.005$ , based on the data of 20 sites, in which cores were sampled at the deepest lake location), supporting the use of the parameter to characterize sediment focusing in the lakes. Each characteristic was statistically standardised to have a zero mean and unit standard deviation in the set of all lakes. This approach generates principal components (PCs) as new complex (synthetic) uncorrelated factors that integrate individual characteristics. This approach is justified by the coexistence of different factors (lake characteristics) that correlate with each other. For example, significant positive correlation was found between the AF and  $D_{\max}$  ( $r = 0.541$ ,  $p = 0.006$ ),  $TP_{in}$  and CA/LA ( $r = 0.612$ ,  $p = 0.002$ ), OI and AF ( $r = 0.509$ ,  $p = 0.011$ ). The effects of the PCs on the  $TP_{acc}$  were estimated by using the general multiparametrical linear model (SAS GLM procedure, type III). Initially, all nine PCs were used together as predictors of the  $TP_{acc}$  to ascertain *significant PCs*. After that, significant PCs were used singly as the predictors of the  $TP_{acc}$ . The significance was adjusted with the Bonferroni's correction. The approach of using PCs as independent variables has proven to be an effective tool in predicting internal P loading and water quality variables (Tammeorg et al. 2017). **Noteworthy, the effect (significant or not) of a PC on TP accumulation does not depend on the proportion of the lake variance that the specific PC describes.**

The correlations between NAO and lake water temperature in the surface and near-bottom water layer, their difference, and wind speed were presented with Pearson correlation coefficient. False discovery rate was applied to multiple testing ( $Q=0.25$  was set as the proportion of the rejected null hypotheses which are erroneously rejected; Benjamini and Hochberg 1995). General multiparametrical linear model was used also to elucidate the effect of NAO and water temperature difference between the surface and near-bottom water layer on TP retention that remained after separating the lake specific effects encompassed under the significant PCs. The  $TP_{acc}$  values were log-transformed to make data distribution close to normal. Statistical analyses were done with SAS (version 9.2, SAS Institute Inc.).

### 3 Results

#### 3.1. Variability across sediment cores, and lake water TP concentrations during two-three decades

The majority of the study lakes had well-defined peaks in the  $^{137}Cs$  activity versus depth records that were confidently attributed to the fallout from the 1986 Chernobyl accident. The good resolution of the Chernobyl peaks suggests that sediment mixing has not been significant and that the  $^{210}Pb$  and  $^{137}Cs$  fallout records stored in the



sediments of these lakes, and the sediment accumulation rates (SARs) determined from those records (Tables S1 – S21), are reasonably reliable. Focusing factors (FFs) were less than two at twelve sites, greater than three at three sites, while intermediate values were found for the remaining six locations (Table 1). The mean FF for all 21 cores studied was about two. Six cores had long-term records spanning periods of time ranging from around 60 years (Kajaanselkä, Äimäjärvi) to more than 120 years (Hormajärvi, Punelia, Puujärvi). The remaining 15 had much shorter records, ranging from 46 years (Tuusulanjärvi) to as few as 19 years (Enonselkä). Mean post-1986 sedimentation rates varied widely from  $0.021 \text{ g cm}^{-2} \text{ y}^{-1}$  (Lake Punelia) to  $0.36 \text{ g cm}^{-2} \text{ y}^{-1}$  (Villikkalanjärvi), being generally higher in the lakes of higher trophic ( $R^2 = 0.56, p < 0.0001$ ; Fig. 1). At twelve sites, the SAR was relatively constant over the last 30 years. Mean SARs at these sites varied by more than an order of magnitude, from  $0.021 \text{ g cm}^{-2} \text{ y}^{-1}$  (Punelia) to  $0.30 \text{ g cm}^{-2} \text{ y}^{-1}$  (Tiiläänjärvi). Since 1986, there were mainly systematic increases in the SAR at eight sites, and decrease in one site (Pusulanjärvi).

In the lakes studied, the mean post-1986 sediment TP concentrations ( $\text{TP}_{\text{sed}}$ ) varied from  $1.1 \text{ mg g}^{-1}$  (Villikkalanjärvi) to  $6.0 \text{ mg g}^{-1}$  (Rehtijärvi).  $\text{TP}_{\text{sed}}$  increased significantly ( $p < 0.01$ ) towards the surface of the core (the most recent years) in nine of the lakes studied that were mainly eutrophic (Table 2). There were increases in  $\text{TP}_{\text{sed}}$  in the lakes with increased SARs (Kynäröjärvi, Loppijärvi, Tuusulanjärvi), with constant SARs (Bodominjärvi, Punelia, Puujärvi, Sahajärvi, Karhujärvi), and in Pusulanjärvi that displayed a decrease in SAR. In Rehtijärvi ( $R^2 = 0.380, p < 0.0001$ ),  $\text{TP}_{\text{sed}}$  decreased over the time period of 30 years. In overall, trends in SAR and  $\text{TP}_{\text{sed}}$  coincided at nine sites. Finally,  $\text{TP}_{\text{acc}}$  increased significantly ( $p < 0.01$ ) in 13 of the lakes (Table 2), decreased ( $p < 0.05$ ) in one lake (Rehtijärvi), while showing no changes in the rest of the lakes (mean for the years since 1986 varied from 340 in Punelia to  $6038 \text{ mg m}^{-2} \text{ y}^{-1}$  in Pusulanjärvi; Fig. 2). At seven sites, trends observed in  $\text{TP}_{\text{acc}}$  coincided with those of the lake water TP concentration (TP), showing no change (Enäjärvi, Pusulanjärvi, and Pyhäjärvi(O)), and increases (Karhujärvi, Villikkalanjärvi, Loppijärvi, and Hormajärvi). At the rest six sites with increased  $\text{TP}_{\text{acc}}$ , TP either decreased (Kajaanselkä, basin of Lake Vesijärvi, Puujärvi, Tuusulanjärvi) or showed no changes (Bodominjärvi, Punelia, Sahajärvi) over the 30-year period.

## 3.2 Factors behind the variability in TP accumulation

### 3.2.1 Lake specifics

The first six and PC8 were found to have significant effect on the  $\text{TP}_{\text{acc}}$ , describing together 60% of the variability of  $\text{TP}_{\text{acc}}$  ( $p < 0.0001$ ). Significant PCs represented about 98% of lake data variability in total (Table 3), whereby

the most of the lakes studied belonged to the types that were characterised by PC1 (39%), PC2 (27%), and PC3 (18%). The effect of PC1 – PC5 on  $TP_{acc}$  remained still significant, when these were used as predictors in the simple linear model (Table 4). The PC1 was mainly representative of the deep lakes (Table 3). In PC2, the highest loadings were by OI (0.516), LA (–0.458), and AF (0.420). By importance for PC3, CA (0.627) was followed by  $TP_{in}$  (0.504) and CA/LA (0.479). The OI (0.654), AF (–0.569), and  $D_{max}/D$  (0.402) were the major constituents of the PC4. The major contributing lake characteristics to PC5 included  $TP_{in}$  (0.620), followed by CA/LA (–0.363) and AF (0.357). In PC6, the highest loadings were by LA (0.612) and  $D_{max}/D$  (–0.570). The PC8 was primarily determined by CA (–0.607), CA/LA (0.564), and LA (0.516). PC6 and PC8 were not significant drivers of  $TP_{acc}$  ( $R^2 = 0.025$ ,  $p = 0.070$ ;  $R^2 = 0.011$ ,  $p = 0.679$ ). In general,  $TP_{acc}$  increased gradually with an increase in the values of the PC1 – PC 5 (Table 4; Fig. 3).

### 3.2.2 Climatic factors

During the years represented in sediment cores, long-term monthly NAO values varied from -1.024 to 1.092 on average (mean values close to zero), whereby somewhat lower values were observed in October (Fig. 4a). Daily mean wind speed was particularly variable during the winter months. Generally, the values decreased towards August, and increased since then (Fig. 4b). The water temperature difference between the surface and bottom layers increased towards July (Fig. 4c), when the highest temperatures reached 20.2 and 13.8 °C in surface water layer and near-bottom water layer, respectively. The temperatures decreased during the following months. As a result, temperature difference between the surface and the near bottom water layer was close to zero in September, October and November (mean values for the corresponding months were 1.2, 0.5 and -0.2 °C).

Monthly NAO correlated significantly with water temperature in the surface and near bottom layer, their difference, and wind speed throughout the year (Table 5). There were many, mainly positive significant correlations of the studied (hydro)meteorological variables with winter-spring NAO and considerably less, mainly negative correlations with the NAO in summer months. Further, a number of (mainly positive) correlations with the NAO increased again in the autumn months. Indeed, reported correlations had the highest significance level mainly in winter-spring, though correlations in winter were as high as in autumn.

Mean NAO value in October showed a potentially significant effect on the  $TP_{acc}$ , additional to those ascribed to the significant PCs, increasing predictive ability of the model ( $R^2 = 0.613$ ,  $p < 0.0001$ ). An increase in NAO index in October by one unit decreased  $TP_{acc}$  1.2 times (19%; Fig. 5). Moreover, the mean NAO index in

October correlated significantly positively with the mean surface and bottom water temperatures and their difference in November ( $r = 0.300$ ,  $p < 0.01$ ;  $r = 0.296$ ,  $p < 0.01$ ;  $r = 0.281$ ,  $p < 0.01$ , respectively; Table 4). The positive effect of the NAO in October on the temperature difference between surface and bottom water layer in November still remained significant (Fig. 5;  $p < 0.01$ ) when the effects associated with the lake specifics were accounted for. At the same time, no significant correlations were found between NAO in October and average wind speed. Average temperature difference in November was  $-0.2^{\circ}\text{C}$  (Fig. 4b), being lower in the surface layer than in the near bottom water layer.

## 4 Discussion

### 4.1 Variations in TP accumulation and its importance

Our results confirm the high importance of the lake trophic state in regulating rates of net sedimentation, one of the determinants of the P accumulation, reported earlier (e.g., Trolle et al. 2009), as these were considerably higher for eutrophic than for mesotrophic lakes. Thus, in case there are no external loading data with sufficient resolution available for a particular lake, changes in net sedimentation rate could shed light on its trophic state history. Our data showed that in most lakes both the sedimentation rates and water TP concentrations either increased or remained constant on the long-term scale, while a decrease in sediment accumulation rate was very rare among the studied lakes. These observations agree with the water quality monitoring data for twenty years in multiple agricultural Finnish lakes showing no improvement in the lake water quality (based on the chlorophyll *a* concentrations; Ekholm and Mitikka 2006).

Changes in trophic state during recent years can possibly explain an increase in sediment TP concentrations in the lakes that displayed also an increase in net sedimentation rate over the recent 30 years (e.g., Loppijärvi, Kynäsjärvi). An increase in TP concentrations over the 30-year period (higher concentrations in the topmost sediments) in lakes with constant sedimentation rates (Bodominjärvi, Sahajärvi, Karhujärvi) is most likely due to diagenetic processes (Carignan and Flett 1981; Trolle et al. 2011). In general, we observed such patterns of sediment TP concentrations mainly in eutrophic lakes. Similarly, elevated concentrations in the surficial sediments representing a large pool of recyclable P were associated with lake eutrophic conditions shown by earlier studies (Carey and Rydin 2011). Moreover, it was not a surprise to observe such a phenomenon in mesotrophic lakes (Punelia and Puujärvi), as the sediments in these lakes can have limited P binding capacity (Carey and Rydin 2011;

Dittrich et al. 2013). Although an opposite vertical distribution of TP concentrations with higher levels in deeper sediments would be expected for oligotrophic lakes (due to Al availability; Carey and Rydin 2011), we found such TP distribution in one highly eutrophic lake (Rehtijärvi). It can be due to a combined effect of post-depositional migration and release of P into water column during periods of anoxia, similarly to what was concluded by Dillon and Evans (1993). This agrees with an increase of the lake water TP concentration in this lake on the long-term scale.

Increases in P could, indeed, result from sediment focusing. This could be of particular concern in cases of Pyhäjärvi (S), Ormajärvi, and the Enonselkä basin of Lake Vesijärvi that had FFs greater than three. However, none of those cores displayed an increase in  $TP_{acc}$ . Moreover, the mean FF for the study area indicates a rather modest level of bias associated with sediment focusing (Heathcote and Downing 2014). Moreover, the  $TP_{acc}$  found in our lakes of mesotrophic and higher trophic state were within the range of the values reported for other lakes of the northern temperate zone (summarized in Tammeorg et al. (2017)), being considerably higher than the values reported for oligotrophic lakes (Dillon and Evans 1993). This increase in  $TP_{acc}$  across the trophic gradient provides a support for the accuracy of our estimates. Therefore, the variations in  $TP_{acc}$  can be explained to considerable extent by differences in lake trophic state, which is closely coupled to lake morphology (Søndergaard et al. 2003; Hupfer and Lewandowski 2008).

The changes in lake water TP concentration similar to those in  $TP_{acc}$  were expected, confirming the high importance of sediments in P budget of lakes (Hupfer and Lewandowski 2008; Søndergaard et al. 2013). There were some lakes in which increased  $TP_{acc}$  appeared to sustain or augment eutrophication (e.g., Villikkalanjärvi, Loppijärvi, Karhujärvi). Unchanged lake TP concentration in lakes that showed an increase in  $TP_{acc}$  can be also due to possible time lags, a well-known phenomenon (Jeppesen et al. 2005). Moreover, restoration efforts could also have a role. In Tuusulanjärvi, in which increased  $TP_{acc}$  co-occurred with decreased lake water TP concentration, food web management applied since 1998 has compensated for the amplified P-cycling, revealed by the decreasing chlorophyll:total P ratio (Horppila et al. 2017).

#### **4.2 Morphometric factors behind variations in TP accumulation**

From the multiple external and internal factors controlling TP accumulation on the long-term scale, our model did not consider those that are related to sediment composition. Nevertheless, the simple model based on lake parameters that are usually available could explain a bulk of the variability in  $TP_{acc}$ . Similarly, there are numerous studies that have shown the association of P retention with morphometric/ hydraulic characteristics of lakes (e.g.,

Vollenweider 1975; Nürnberg 1984; Dillon and Molot 1996; Brett and Benjamin 2008; Kõiv et al. 2011). Generally,  $TP_{in}$  and hydraulic retention time are of key role in regulating TP retention in the classical models. Our model takes into account the additional characteristics that classical models lack, i.e. the anoxic factor reflecting P release and the Osgood's index characterising water column stability (Nürnberg 1984; Nürnberg 2009). Moreover, while P retention is conventionally calculated as a coefficient from mass balance equation (Brett and Benjamin 2008), we connected observed P accumulation rates with lake specific features. Interestingly, despite being quite different, our model gave nearly identical results (similar  $R^2$ ) to classical ones. Similarly, Benjamin and Brett (2008) concluded that various multiple regression models yield very similar fits to those of Vollenweider type analyses because the terms typically considered in share many variables. While this makes the use of more simple models more preferable, we claim the approach used here to better take into account lake specifics via the use of PCs as independent factors. The relevance of the approach is supported, for example, by the finding of Kõiv et al. (2011) who showed that the retention is much more strongly determined by external P loading and hydrological residence time in large lakes than in smaller lakes.

Although the PCs represent a combination of different lake characteristics, they are somewhat predetermined by the values of some particular drivers (main contributors). Lake depth ( $D$ ,  $D_{max}$ ), the main contributor to the PC1, has been generally acknowledged as a factor that favours P accumulation (Håkanson and Jansson 1983), which agrees with the positive effect of PC1 on  $TP_{acc}$  in our study. However, the largest proportion of  $TP_{acc}$  variability was ascribed to changes in PC2, PC3, and PC5. The P accumulation appeared to be high in small lakes, with high water column stability, and high anoxic factor. The sediment P pool is often small in large lakes because resuspension leads to washout of particulate TP and organic net sedimentation is low, the latter due to high mineralization (Jeppesen et al. 2007). In small lakes, conditions are more favourable for stable stratification, and the relative importance of anaerobic areas is high. Both PC3 and PC5 characterize productive lakes, as  $TP_{in}$  is one of the major constituents of those components. In general, high  $TP_{in}$  results in the increased deposition of newly-produced P-rich material (Marsden 1989; Carey and Rydin 2011). The productivity is associated with large CA and high CA/LA values in the PC3, while with low CA/LA and relatively high AF in the PC5, suggesting differences in the relative importance of internal and external P loading in lakes. Lower CA/LA values are indicative of longer residence times and higher percentage of P load from internal sources with strong implications of the sediment P sources for productivity and water quality (Huser et al. 2016). High levels of external loading often result in oxygen deficits that sustain the recycling of P to the water column (Gächter and Wehrli 1998; Moosman et al. 2006) through the breakdown of the iron-phosphorus complexes (Einsele 1936;

Mortimer 1941, 1942). On the other hand, PC3 is likely to represent the lakes with productivity determined by external P sources. Allochthonous, mineral-bound particulate matter is more prone to settling, resulting in higher loss of P in lakes with shorter residence time (Brett and Benjamin 2008). Finally, TP accumulation tended to be high in lakes with high water column stability, but low anoxic factor, and relatively high  $D_{\max}/D$  characterizing sediment focusing (PC4).

#### **4.3 Climatic variability as a potential factor behind temporal changes in TP accumulation**

One of the most interesting findings of the current study is that NAO in October influenced potentially the annual  $TP_{acc}$ , explaining its variability in addition to the significant PCs. Previously, the most pronounced implications for lake ecosystems were ascribed to the NAO values in winter-early spring (e.g., Bleckner et al. 2007; Pettersson et al. 2010; Spears and Jones 2010). Similarly, we found numerous significant correlations of NAO with wind speed and water temperatures at this period of time. While interpreting the results, it should be considered that NAO affects simultaneously air temperature, precipitation, wind speed and direction, cloudiness etc., each of them having potential feedbacks to lakes involving different lag periods. In the study region, the mechanisms behind NAO effects on the lakes functioning are mostly related with hydrology and ice regime, as milder temperatures cause more thaw days with increased runoff and shorter duration of ice cover (Nöges et al. 2010; Pettersson et al. 2010). Moisture transported from North-Atlantic causes more precipitation that acts in the same direction increasing the runoff (Hurrell and Van Loon 1997). Also in southern Finland, precipitation in winter was found to strongly associate with NAO (Irannezhad et al. 2014). Increased runoff mostly increases nutrient loading, if available in the catchment (Jeppesen et al. 2009; Trolle et al. 2011), entailing an increase in  $TP_{acc}$ . However, also opposite effect can be expected by flushing with meltwater, which is very likely to occur in the studied lakes with small area and depth. Nevertheless, our study revealed a potential importance of the autumnal NAO values for the water temperatures between the surface and near-bottom water layer, and their difference suggesting therefore possible mechanisms behind the changes in  $TP_{acc}$ .

The changes in the water temperature difference are most likely linked to NAO via air temperature. This suggestion is supported by the significant positive correlation between NAO and air temperatures in autumn reported for Finland (Irannezhad et al. 2015). Although there are no equivalent data reported for the wind, the results obtained for the nearby areas suggest its relevance as a possible explanatory mechanism from the perspective of both temperature difference and TP retention. Vermaat et al. (2008) showed that a positive NAO leads to increased wind-induced turbulence, and hence to higher resuspension of particle-bound nutrients.

Similarly, Spears and Jones (2010) showed that positive NAO correlated with stronger, more westerly winds, though correlations (including correlation between NAO and wave-mixed depth) were found only for winter and spring. Nevertheless, our data for Finnish lakes did not reveal any significant correlations of autumnal NAO with wind speed during September–November, suggesting the key role of air temperature in regulating autumnal water temperatures and a difference in temperature, which are likely to be linked to  $TP_{acc}$ .

Climate warming is generally associated with prolonged stratification in lakes leading to prolonged periods of anoxia, and release of P from sediments (Jeppesen et al. 2009). Similarly, Snorheim et al. (2017) found significant positive correlations between air temperature and anoxic factor for the northern dimictic Lake Mendota, additionally showing that the factor had the greatest potential impact for the stratification conditions (from the other studied factors, as wind speed and humidity). However, our water temperature data showed that stratification can be broken already since September, and water temperature difference in November (potentially linked to decreased TP retention) is negligible. Moreover, the prolonged algal blooms, associated with higher temperatures, are expected to result in higher supply of the organic matter and associated nutrients to the sediment during autumn (Blenckner et al. 2007; Jeppesen et al. 2009; Trolle et al. 2011). Therefore, this scenario suggests an importance of additional drivers to explain reduced  $TP_{acc}$ , e.g., flushing. Previously, a significant positive correlation was found between NAO and DOC discharge from the River Oulujoki in autumn (Marttila et al. 2014). However, the most likely mechanism seems to be associated with increased mineralization of the organic material in the sediments due to increased temperatures, as concluded by Gudasz et al. (2010). Over the boreal zone, the increase in organic carbon mineralization in sediments overlain by mixed water due to temperature increase (range of 1.8–4 °C) was predicted to result in a decrease of organic carbon burial of 6–15% in lake sediments (Gudasz et al. 2010). Providing that the sediment P is closely related to organic carbon (Håkanson and Jansson 1983), this would be the most likely explanation for the reduced TP sedimentation potentially associated with NAO driven water temperature increase.

## 5 Conclusions

The lake characteristics explained bulk of the variability in TP accumulation ( $TP_{acc}$ ).  $TP_{acc}$  tends to be high in the lakes with following features: 1) mainly deep lakes; 2) small lakes with high levels of anoxia and water column stability; 3) lakes with high levels of P inflow, large catchment area and high CA/LA; 4) lakes with high water column stability, low anoxic factor, and relatively high sediment focusing; 5) lakes with high levels of P inflow,

anoxia and low CA/LA. Additionally to the effects attributed to lake specifics, we found a negative effect of NAO in autumn on annual  $TP_{acc}$  in Finnish lakes. The temperatures in surface and bottom water layer and their difference in autumn in the lakes studied were potentially related with NAO, suggesting the possible implications for P dynamics. Hence the analysis presented here for an internally consistent dataset (sampled in the same way) seems to better take account of lake specifics than previously. Moreover, this spatially and temporally comprehensive sediment data can potentially be a valuable source for modelling climate change implications.

**Acknowledgments.** The research was supported by the Academy of Finland (grant nr. 263365), Maa- ja vesitekniikan tuki ry (grant nr. 29159), and the City of Lahti. We thank Heidi Holmroos, Lauri Happonen, Jani Ruohola, and Raija Mastonen for their help in the field and in the lab. J. Weckström is acknowledged for useful discussions.

**Funding:** This study was funded by the Academy of Finland (grant nr. 263365), Maa- ja vesitekniikan tuki ry (grant nr. 29159), and the City of Lahti.

**Conflict of Interest:** The authors declare that they have no conflict of interest.

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**Table 1.** Basic lake characteristics, including lake trophic status (TI), mean and maximum depth (D, D<sub>max</sub>), catchment area (CA), lake area (LA), anoxic factor (AF) values, mean phosphorus inflow (TP<sub>in</sub>), age of the 20-cm sediment cores sampled in 2014, focusing factor (FF), and restoration activities of the study area. For the AF and TP<sub>in</sub> mean values for the period 1986–2014 are presented.

Lake	Coordinates	TI	D (m)	D <sub>max</sub> (m)	CA (km <sup>2</sup> )	LA (km <sup>2</sup> )	AF d y <sup>-1</sup>	TP <sub>in</sub> (mg P m <sup>-2</sup> y <sup>-1</sup> )	Age y	FF	Restoration measures applied
Äimäjärvi	61°03'N 24°10'E	eutr	2.9	9	93	8.5	22.1	228	65	0.7	Wastewater diversion in 1969, biomanipulation since 1997, sedimentation ponds established in 2004
Bodominjärvi	60°15'N 24°40'E	eutr	4.3	12.7	32	4.1	7.2	507	31	2.5	Chemical treatment, in 1980 oxygen-depleted water diversion, aeration from 1970s to 1998
Enäjärvi	60°20'N 24°22'E	eutr	3.2	9.1	34	4.9	1.8	323	89	0.8	Wastewater diversion in 1976, fish removal from 1993, aerated from 1998
Enonselkä	61°00'N 16°36'E	eutr	6.8	33	84	26	24.7	137	19	3.9	1976-1978 sewage diversion, aeration (since 2009), biomanipulation (1989-1993), sedimentation ponds and wetlands
Hormajärvi	60°17'N 24°01'E	meso	7.3	21	16	5.1	36.1	88	121	1.8	Aeration since 2008
Kajaanselkä	61°09'N 25°28'E	meso	6.8	42	138	44	1.2	124	64	1.9	-
Karhujärvi	60°14'N 24°17'E	eutr	2.2	4.9	142	1.9	0.0	253	25	1.9	Dredging of the shore areas in 1997, fish removal since 1996, measures to reduce macrophyte expansion (1992, 1993, 1994-1995)
Katumajärvi	60°59'N 24°31'E	meso	7.1	18.9	51	3.8	29.4	232	38	2.4	Sedimentation ponds established in 2004-2005, biomanipulation 2003-2005
Kyynäröjärvi	61°07'N 24°59'E	eutr	1.3	3	25	0.25	0.0	2263	42	0.6	-
Loppijärvi	60°41'N 24°25'E	eutr	1.8	6.7	82	11.8	0.0	93	34	1.4	Wastewater diversion in 1975, wetlands and sedimentation ponds, fish removal since 1990s
Ormajärvi	61°06'N 24°59'E	meso	9.6	29.4	86	6.6	48.6	128	31	3.3	-
Punelia	60°41'N 24°12'E	meso	3.8	14	102	6.8	12.7	23	126	2.4	-
Pusulanjärvi	60°27'N 23°59'E	eutr	4.9	10.6	226	2.1	49.7	1707	35	1.6	Wastewater load until 1988, intensive fishing, Aeration (first in 1989)
Puujärvi	60°15'N 23°43'E	meso	8.3	21.7	27	6.4	15.0	26	173	1.7	-
Pyhäjärvi (A)	60°43'N 26°00'E	eutr	21	68	459	12.9	32.9	814	34	2.3	-
Pyhäjärvi (S)	61°00'N 22°18'E	meso	5.5	26.2	461	155	4.5	106	27	4.5	Intensive commercial fishing (+ fish removal)
Rehtijärvi	60°51'N 23°29'E	eutr	9.2	30	3	0.4	47.3	628	29	1.0	Wetlands and sedimentation ponds in 1994-1998, biomanipulation
Sahajärvi	60°44'N 25°28'E	eutr	4.3	11	26	1.92	26.2	403	28	2.3	-
Tiiläänjärvi	60°32'N 25°42'E	eutr	4.4	10.3	38	2.1	26.9	1428	25	2.2	-
Tuusulanjärvi	60°25'N 25°04'E	hyper	3.2	10	92	5.9	26.5	960	46	1.1	Wastewater diversion since 1979, aeration since 1970, wetlands and biomanipulation since 1998
Villikkalanjärvi	60°47'N 26°02'E	hyper	3.2	10	413	7.1	18.5	2302	27	1.5	Fish removal

**Table 2.** Sediment accumulation rate (SAR), sediment phosphorus concentration (TP<sub>sed</sub>), phosphorus accumulation rate (TP<sub>acc</sub>), and TP concentration of the lake water (TP) in lakes studied for 1986–2014. Significant trends in SAR, TP<sub>sed</sub>, TP<sub>acc</sub>, and TP (increase “+”, decrease “-”) over the 30-year period are shown (T1, T2, T3, and T4, respectively), and “0” indicates no significant changes. Long-term data on water quality were not available for Lake Kyyjärvi (indicated as “na”).

Lake	SAR (g cm <sup>-2</sup> y <sup>-1</sup> )	T 1	TP <sub>sed</sub> (mg g <sup>-1</sup> )	T 2	TP <sub>acc</sub> (mg P m <sup>-2</sup> y <sup>-1</sup> )	T3	TP (µg l <sup>-1</sup> )	T4
Aimajärvi	0.04	+	2.2	0	907	+	42	-
Bodominjärvi	0.16	0	1.5	+	2373	+	32	0
Enäjärvi	0.04	0	2.7	0	1074	0	98	0
Enonselkä	0.18	0	2.7	0	4910	0	34	-
Hormajärvi	0.03	+	5.3	0	1407	+	14	+
Kajaanselkä	0.08	+	1.6	0	1418	+	16	-
Karhujärvi	0.15	0	1.2	+	1827	+	72	+
Katumajärvi	0.06	+	4.6	0	2610	+	19	0
Kyyjärvi	0.16	+	1.6	+	2540	+	50	na
Loppijärvi	0.07	+	2.3	+	1699	+	30	+
Ormajärvi	0.06	0	3.6	0	2073	0	20	-
Punelia	0.02	0	1.8	+	340	+	14	0
Pusulanjärvi	0.19	-	3.2	+	6038	0	49	0
Puujärvi	0.02	0	2.2	+	508	+	12	-
Pyhäjärvi (A)	0.17	0	2.1	0	3502	0	44	0
Pyhäjärvi (S)	0.11	0	2.3	0	2488	0	17	+
Rehtijärvi	0.20	0	6.0	-	12044	-	56	+
Sahajärvi	0.17	0	1.7	+	2906	+	40	0
Tiiläänjärvi	0.30	0	1.5	0	4533	0	80	+
Tuusulanjärvi	0.11	+	1.7	+	1894	+	101	-
Villikkalanjärvi	0.36	+	1.1	-	4097	+	111	+

**Table 3.** Coefficients for calculating the first six (of eight) principal components (PCs) and corresponding eigenvalues of the correlation matrix calculated for 21 lakes with a full set of all eight contributing characteristics. The characteristics needed include: maximum depth ( $D_{\max}$ ), lake area (LA), catchment area (CA), mean depth (D), ratio of the CA to LA, inflow of P ( $TP_{in}$ ), anoxic factor (AF, defined as the product of the duration of anoxia and the percentage of the anaerobic areas), Osgood's index, or  $D \times LA^{-0.5}$  (OI).

Lake	PC 1	PC 2	PC 3	PC 4	PC 5	PC 6	PC 8
Characteristics							
D	0.441	0.265	0.201	-0.101	-0.334	-0.033	0.076
$D_{\max}$	0.500	0.105	0.177	0.102	-0.174	-0.222	0.130
CA	0.072	-0.334	0.627	-0.097	-0.217	0.164	-0.607
LA	0.318	-0.458	0.113	0.033	0.176	0.613	0.516
AF	0.263	0.420	0.195	-0.569	0.357	-0.056	0.107
CA/LA	-0.374	0.032	0.479	0.177	-0.363	-0.265	0.554
OI	0.157	0.516	-0.002	0.653	-0.040	0.383	-0.150
$TP_{in}$	-0.270	0.195	0.504	0.163	0.620	0.042	-0.004
$D_{\max}/D$	0.376	-0.339	0.015	0.402	0.367	-0.570	0.044
Eigenvalue	3.480	2.414	1.636	0.493	0.470	0.228	0.079
Proportion of variability	0.387	0.268	0.182	0.057	0.055	0.052	0.009
Cumulative proportion	0.387	0.655	0.837	0.892	0.944	0.969	0.978



**Table 4.** Most significant predictors of the phosphorus accumulation rate (log-transformed values) according to the linear model. The effects indicate the change of the phosphorus accumulation rate when significant principal component (PC) changes by one unit.

Significant predictors	Effect	$R^2$	$p$
PC 1	$2.78 \pm 0.82$	0.044	0.006
PC 2	$5.86 \pm 0.75$	0.197	< 0.0001
PC 3	$4.88 \pm 0.78$	0.137	< 0.0001
PC 4	$4.95 \pm 0.76$	0.046	0.004
PC 5	$6.17 \pm 0.85$	0.141	< 0.0001

**Table 5.** Significant correlations (indicated by Pearson correlation coefficient) of monthly NAO values with monthly values for the (hydro)meteorological variables including water temperature in the surface layer, near bottom water layer and difference in these water temperatures.

Month		NAO_1	NAO_2	NAO_3	NAO_4	NAO_5	NAO_6	NAO_7	NAO_8	NAO_9	NAO_10	NAO_11	NAO_12
Jan	bot	0.210*											
	wind	0.601****											
Feb	bot	0.197*											
	wind		0.436*										
Mar	surf		0.271****										
	dif		0.142**	0.178***									
	wind			0.337*									
Apr	surf		0.216*	0.485****									
	bot			0.223*									
	dif		0.250*	0.403****									
May	bot		0.163**	0.152*									
	dif		-0.189**										
	wind	-0.347*											
Jun	surf					0.191*							
	dif					0.208**							
Jul	surf			-0.203***									
	bot							0.326*					
	dif			-0.144*									
Aug	surf				-0.227****								
Sep	surf			-0.190*									
	wind		-0.345*										
Oct	bot								-0.180**				
	dif								0.170**				
	wind				-0.310*				-0.427**				
Nov	surf									0.291**	0.300**		
	bot									0.264**	0.256**		
	dif									-0.218*	0.229**	0.281**	
	wind		-0.365*	-0.321*									
Dec	surf								-0.353*				
	bot			0.314*									
	dif			-0.413**				-0.434**					
	wind											0.325*	0.426**

\*\*\*\* - the level of significance  $p < 0.0001$ ; \*\*\* -  $p < 0.001$ ; \*\* -  $p < 0.01$ ; \* -  $p < 0.05$ .

## Figure Captions

**Fig. 1** Variations of the sediment accumulation rate in mesotrophic (meso), eutrophic (eutr) and hypertrophic (hyper) lakes of southern Finland

**Fig. 2.** Phosphorus accumulation rate ( $TP_{acc}$ ) in 21 Finnish lakes over 1986–2014.

**Fig. 3.** Dependence of the phosphorus accumulation rate (log-transformed values) on the specific combination of lake characteristics represented by PC5 that characterizes mainly productive lakes

**Fig. 4.** Monthly variations in NAO (a), daily mean wind speed (b), and water temperature differences between surface and near bottom layers of the lakes studied (c) during 1986–2014.

**Fig. 5.** Dependence of the annual phosphorus accumulation rate ( $TP_{acc}$ ;  $\log(TP_{acc}) = 7.4 - 0.4 * \text{NAO in October}$ ), and water temperature difference between the surface and near bottom layer in November (temperature difference in November =  $0.4 * \text{NAO in October} - 0.06$ ) on the NAO in October